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Annealing and morphology transformation effects in MOCVD grown of self-organized InAlAs-AlGaAs quantum dots

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Abstract. We report on MOCVD growth of the (In,Al)As-self-organized quantum dots (QD) on (Al,Ga)As. We demonstrate that dense arrays ($\sim 2 \times 10^{10}$ cm^{-2}) of small ($\sim$ 5 nm) QD are formed during annealing (750°C, 20 min, excess of arsine) of $In_{0.5}Al_{0.5}As$ deposited at 500°C on a $Al_{0.6}Ga_{0.4}As$ surface, as demonstrated by atomic force and transmission electron microscopies and by photoluminescense. We determined that the ripening process continues at room temperature and gives rise to large clusters of submicron size after time of one. In contrast, an identical sample that is not annealed shows low density (2×10^9 cm^{-2}) of the QDs. Aging in this case gives results in a gradual smoothing of the morphology.

Introduction

The self-organized growth of semiconductor zero-dimensional structures usually includes the direct formation of quantum dots (QDs) via the Stranski–Krastanow growth mode. At the same time it has been demonstrated that Ostwald ripening process [1, 2, 3], resulting in an increase of the QD volume and shape transformations from pyramids to domes, can occur during annealing [4, 5, 6, 7] or even at room temperature for some material systems [8]. Here we study and observed similar effects for InAlAs/AlGaAs QDs. Structures in this system have been previously grown by MBE [9, 10]. Here we fabricate and characterize self-organized InAlAs-AlGaAs quantum dots using the MOCVD growth method and report pronounced morphological transformation effects induced in this system by annealing.

1 Experiment

The heterostructures were grown using equipment with a horizontal resistively heated reactor at low pressure (76 torr). Trimethylgallium, trimethylaluminium, ethyldimethylindium, and arsine were used as the sources of materials. The ratio of the group V and group III elements was 75. The growth was carried out with an excess of arsine. Si-doped (100) GaAs substrates were used. Three type of structures were grown. In all cases, after the GaAs buffer-layer growth, a 0.5 μm-thick AlAs layer was deposited and followed by 0.3 μm thick $Al_{0.6}Ga_{0.4}As$ layer. Then a sheet of InAlAs QDs was deposited at 500°C. The average thickness of deposited InAlAs was 2.5 nm and the average In composition was 0.5.

In the one type structure (designated as type A) after the deposition of InAlAs QDs the temperature was increased from 500 to 750°C and growth was interrupted on 20 min. After such *in situ* annealing the 0.1 μm thick $Al_{0.6}Ga_{0.4}As$ layer was deposited at 750°C. In the another structure (type B) no growth interruption occured, and a 0.1 μm thick $Al_{0.6}Ga_{0.4}As$

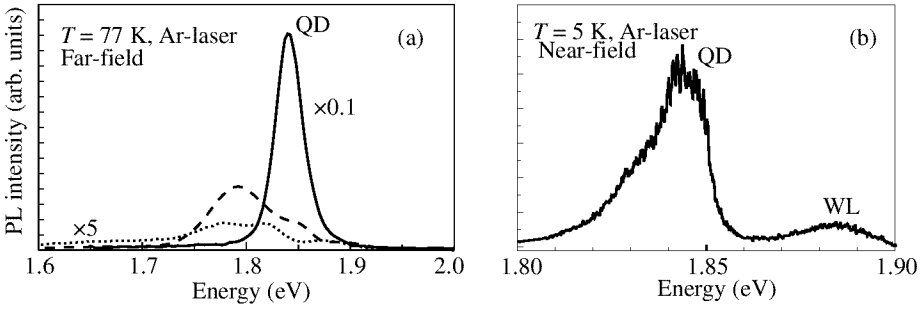

Fig. 1. PL spectra of InAlAs QDs, (a) (far-field): solid curve—structure A, dotted and dashed curves—non-annealed and annealed structure B; (b) (near-field)—structure A.

cap layer was deposited at 500°C. Structures of type A and B were used for PL and TEM measurements.

A third structure (type C) used for AFM measurements had no AlGaAs cap layer. Samples consisting of structure B and C were annealed at 750°C during 20 minutes.

The PL spectra were taken at 77 K under the using 5 mW excitation of the 514.5 nm Ar-laser line. Near-field scanning optical microscope (NSOM) measurements were done at 5 K in illumination mode as described in [11].

2 Results and discussion

2.1 PL measurements

The PL spectra of the structure A and structure B (as grown and annealed) are shown in Fig. 1(a). For the structure A we can see only one very strong band (QD). The relativley small PL linewidth (30 meV) indicates a large size uniformity of the resulting QDs. In Fig. 1(b) we show a NSOM spectrum of the same luminescense line. It is clearly seen that the line consists of multiple ultranarrow peaks with the narrowest ones having a sub-meV halfwidth.

In contrast spectra of structure B contain several broad bands, having very low PL intensity, indicating low density and uniformity of QDs [12]. Annealing of sample B leads to an increase and a redistribution of the PL intensity, and to a blue shift of the bands. This suggests a changing in the island size and in In composition, probably via partial interdiffusion of In and Al [13].

2.2 TEM studies

A cross sectionTEM image of structure A is presented in Fig. 2. The TEM measurements clearly reveal strain-induced contrast characteristic of QDs and point to a typical dot sizes of about 15–20 nm and relative separation between islands of about 40–50 nm. The estimated area density is about 4×10^{10} cm^{-2}.

2.3 AFM measurements

In Fig. 3(a,b) we present the surface morphology of non-annealed and annealed sample C. Self-organized islands are clearly seen in the images. For the as grown sample (Fig. 3(a)) the islands have low density (2×10^9 cm^{-2}). The dominant islands are $\sim$ 40 nm (apparent size $\sim$ 50 nm in AFM image) size islands, while smaller islands of $\sim$ 15 nm ($\sim$ 25 nm in AFM image) size are also presented. After annealing (Fig. 3(b)) the islands density dramatically increased up to 2×10^{10} cm^{-2} and the $\sim$ 15 nm size islands become dominant.

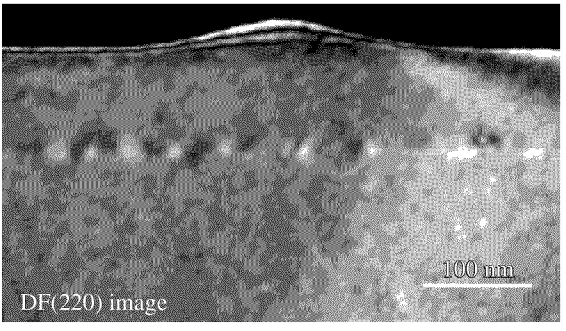

Fig. 2. Cross section TEM image of InAlAs QD structure

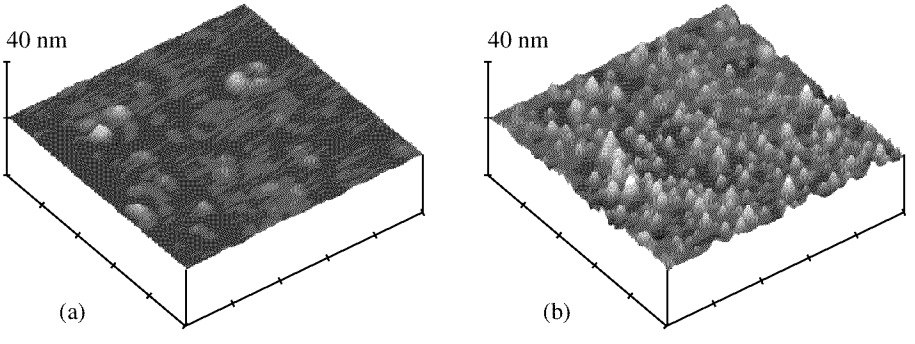

Fig. 3. Surface morphology of as grown (a) and annealed (b) sample C. Scan area 1×1 μm.

In Fig. 4(a,b) we present the surface morphology of the same samples remeasured one year later. No islands are seen in the non-annealed sample. In the annealed sample practically all small islands disappear and big islands (domes) with base ~ 0.3 μm (height 60 nm) and ~ 0.5 μm (height 150 nm) appear.

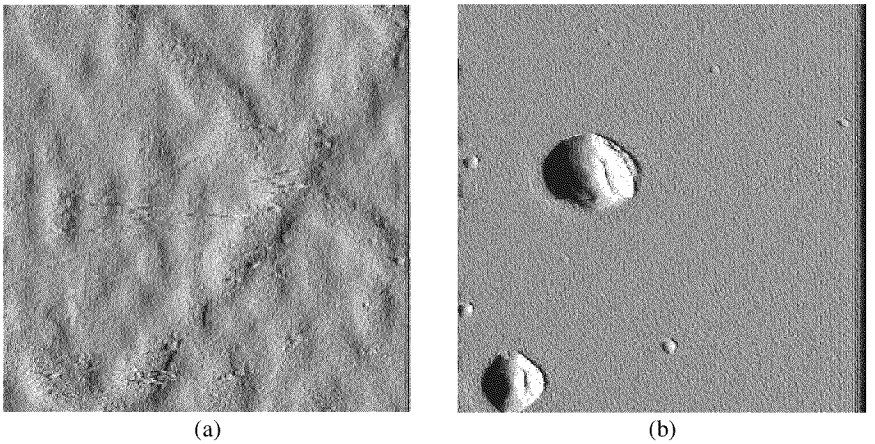

Fig. 4. Surface morphology of non-annealed (a) and annealed (b) sample C, remeasured after one year. Scan area 2×2 μm.

2.4 Discussion

It must first be emphasized that in the annealed samples the structural parameters of the islands, their density, the thickness of the wetting layer are all close to those properties reported for MBE growth [9]. Second, we demonstrate that InAlAs islands obtained via annealing are unstable at room temperature and transform into large clusters as a function of time due to Ostwald ripening. Third, we shown that annealing plays a dominant role in the formation of dense arrays of InAlAs islands by MOCVD. This can be explained as the overgrowth of the wetting layer, delaying the onset of island formation [14]. Upon annealing, such an overgrown wetting layer should lose most of excess material, with atomic rearranment to form strained islands as they tend toward equilibrium. On the other hand, the delay of the onset of island formation can be connected with changing of the value of the critical thickness for 2D-3D growth mode transition. This point of view must assume that as grown InAlAs film has strong non-uniformity of In distribution and annealing changes the distribution of In via diffusion. In this case one may expect the difference in the time evolution of the surface morphology of non-annealed and annealed films which has been experimentally observed.

References

[1] J. Tersoff and F. K. LeGoues, *Phys. Rev. Lett.* **72**, 3570 (1998).
[2] V. A. Schukin, et al., *Phys. Rev. Lett.* **75**, 2968 (1995).
[3] I. Drauka and A.-L. Barabashi, *Phys. Rev. Lett.* **79**, 3708 (1997).
[4] F. M. Ross, et al., *Phys. Rev. Lett.* **80**, 984 (1998).
[5] R. Leon et al., *Phys. Rev. Lett.* **81**, 2486 (1998).
[6] G. Mederiros-Ribeiro et al., *Phys. Rev. B* **58**, 3533 (1998).
[7] B. D. Min et al,, *Phys. Rev. B* **57**, 11879 (1998).
[8] S. Lee et al., *Phys. Rev. Lett.* **81**, 3479 (1998).
[9] R. Leon et al., *Appl. Phys. Lett.*. **64**, 521 (1995).
[10] A. F. Tsatsul'nikov et al., *Appl. Surf. Sci.* **23/124**, 381 (1998).
[11] H. D. Robinson, *Appl. Phys. Lett.* **72**, 2081 (1998).
[12] R. Leon and S. Fafard, *Phys. Rew B* **58**, 1726 (1998).
[13] A. O. Kosogov et al., *Appl. Phys. Lett.* **69**, 3072 (1996).
[14] J. K. Furdyna, S. Lee, A.-L. Barabashi and J. L. Merz, "Self-Organized Low-Dimensional II-VI Nanostructures", in *II–VI Semiconductor Materials and Their Applications*, ed. M. C. Tamargo (Gordon and Breach Science Publishers, 1999) (in press).